Polarized neutron reflectometry studies of GaMnAs/GaAs superlattices

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Abstract. Polarized neutron reflectometry has been used to investigate details of spin ordering in ferromagnetic (FM) GaMnAs/GaAs superlattices. The reflectivity spectra measured below the Curie temperature reveal additional magnetic contributions to the structural superlattice Bragg peaks, clearly indicating the existence of FM interlayer correlations. Closer investigation of the magnetic reflectivity maxima using a full polarization analysis provides direct evidence that the FM order in the GaMnAs layers is truly long-range. Moreover, as shown by the data, the system exhibits a strong tendency of forming a single-domain FM arrangement, even when cooled through T_C in zero external field.

Currently, a great deal of attention is being focused on a new area of solid-state electronics, usually referred to as 'spintronics' [1]. In contrast to conventional electronics, in spintronics not only the current magnitude, but also its spin state, is controlled. Spin valves and spin injectors are the first examples of practical applications of spintronics. Further progress in developing new devices hinges critically on the availability of suitable materials. Such materials need to be 'good' semiconductors, easy to integrate in typical ICs, and their semiconductor properties should exhibit strong sensitivity to the electronic spin states. An especially desirable property is ferromagnetism, as it greatly enhances the effect of an external magnetic field. Unfortunately, there are few natural ferromagnetic (FM) semiconductors, and none of them are particularly useful for spintronics applications, due to either low Curie temperatures (e.g. EuO, EuS) or structural incompatibility with materials typically used in semiconductor technology (e.g. $CuCr_2Te_3I$). In search of better materials, since the mid-1970s researchers started investigating systems known as diluted magnetic semiconductors (DMSs), which are derived from canonical semiconductors (e.g. CdTe or GaAs)

by substituting a controlled fraction of non-magnetic cations by magnetic ions (Mn, Fe, Eu, . . .). One well-known DMS family, intensively studied for more than two decades, includes materials based on the II–VI compounds [2]. However, even though the II–VI-based DMS alloys exhibit a range of highly interesting properties, they are not good for spintronics applications because they are all antiferromagnetic. Yet, recent progress in the molecular beam epitaxial growth of III–V-type DMSs (e.g. GaMnAs, InMnAs, GaMnN) [3] raises new hope, since they seem to possess all necessary features of a 'good' spintronics material. In particular, the strongly p-type InMnAs and GaMnAs exhibit the desired FM behavior [4]. The Curie points of these materials are still much below room temperature, which limits their practicality. Nevertheless, they may play an important role in developing prototypes of future spintronic devices.

The most desirable situation from the spintronics viewpoint is spontaneous formation of a single-domain FM state, thus reducing the need for an external magnetic field. Our current insight into the domain structure of the new epitaxial ferromagnets is still insufficient. In general, the present knowledge of GaMnAs ferromagnetism is based on magnetization and transport measurements, which probe only the volume properties of the spin system [4, 5]. Such data do not provide conclusive information about the range of the FM ordering. Insight into this issue can be obtained only by methods capable of probing the magnetic correlations on a microscopic level. In the present communication we report neutron reflectometry data from GaMnAs/GaAs superlattices (SLs). By applying the technique of polarization analysis we were able to observe the magnetic scattering contributions from domain states. The results show that in most samples cooling through the Curie temperature in zero external field leads to the formation of single-domain ferromagnetic order in the multilayered structure. Such behavior clearly indicates that the ferromagnetism occurring in individual GaMnAs layers is truly long-range. Furthermore, the reflectometry data prove that there is a significant exchange coupling between the GaMnAs layers across the intervening non-magnetic GaAs spacers.

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The samples used in the reflectometry measurements were in the form of GaMnAs/GaAs superlattices. Peaks in the neutron reflectivity occur at positions corresponding to the superlattice periodicity, so that scattering from the substrate does not pose a problem. The magnetic and nuclear scattering components in the reflectivity maxima are of comparable intensity, and the latter component can be completely cut off using polarization analysis.

The SL samples for the present study were grown by molecular beam epitaxy on GaAs (001) substrates. The Mn concentration in the GaMnAs layers was 6%. In various samples investigated the GaMnAs layer thickness was 25 or 50 monolayers, and the GaAs spacer thickness was 4, 6 or 8 monolayers. The number of repeats in all samples was 50. The reflecting surface area of the samples was of the order of $1–1.5\,cm^2$. As probed by magnetization measurements, the samples were ferromagnetic below ~ 40 K (the details of the growth procedure and the magnetic characterization are presented elsewhere [6]).

Neutron reflectivity measurements were carried out at the NIST Center for Neutron Research using the NG-1 reflectometer in both polarized and unpolarized operation modes. The neutron wavelength was $\lambda = 4.75$ Å. In the polarized mode all four types of the cross sections, corresponding to spin-flip (SF) and non-spin-flip (NSF) scattering processes, were measured.

Even though 0.06 is close to the maximum attainable Mn concentration, it is still a relatively small value, and the scattering length density (SLD) contrast between the constituent layers is quite weak. Consequently, the Bragg-peak intensity is rather low. Fortunately, polarized neutron techniques offer the possibility of detecting very small magnetic scattering effects superimposed on much higher nuclear ones by taking advantage of the interference between the magnetic and nuclear contributions to the scattering (e.g. [7]). Another great advantage of a polarized neutron beam is its ability to sense the direction of the in-plane layer magnetization, and thus to probe separately different domain populations. As is well known (e.g. [8]) the component of the magnetization parallel to the applied field (vertical in our case) gives rise to NSF scattering, which also includes contributions from the nuclear scattering. The perpendicular component (the horizontal one in our arrangement) is the source of purely magnetic SF scattering.

Table 1 shows the calculated scattering length densities for the constituent materials of the superlattice for the $(++)$ and $(--)$ NSF scattering cross sections and the $(+-)$ SF scattering cross section. Due to the negative value of $b_{Mn} = -3.73$ fm and a positive magnetic contribution from the $\frac{5}{2}$ Mn spin [9], there is almost perfect compensation of the SLD contrast between GaMnAs and GaAs for the $(--)$ NSF scattering cross section. Consequently, for this neutron spin

direction there should be no observable superlattice Bragg peak.

For the other neutron polarization state, $(++)$, the SL peak is enhanced as compared with the unpolarized neutron experiment. Both the $(++)$ and the $(--)$ NSF reflectivity profiles for GaMnAs/GaAs (50 ML/6 ML)×50 are shown in Fig. 1. The absence of the SL Bragg peak in the $(++)$ data and its presence in the $(--)$ data means that the layer moments are aligned in the direction opposite to the applied magnetic field. In Fig. 1 the theoretical reflectivities were determined using the optical treatment of the reflection process from stratified homogeneous media [10], where the total reflectivity of the superlattice is calculated recurrently from the reflection coefficients of each individual interface. Excellent agreement between calculated and experimental reflectivity profiles has been achieved for the SLD values presented in Table 1.

In Fig. 2 we present a closer look at the first-order superlattice Bragg peak. The results of all four NSF and SF scattering processes are displayed for experiments performed below and above T_C and in applied external fields of 2 and 100 G. Above T_C (Fig. 2c) the sample is non-magnetic (paramagnetic), and both the $(++)$ and $(--)$ cross sections coincide since only nuclear scattering is present. There is also no spin-flip scattering. At 7.8 K and in a 100 G external field (Fig. 2b) the sample is saturated and its magnetization is aligned parallel to the magnetic field. There is a SL Bragg peak in the $(++)$ scattering cross section and no such peak in the $(--)$ data, as one should expect according to the values of the SLD presented in Table 1. For the saturated sample one should not expect any spin-flip scattering as there is no magnetization component perpendicular to the applied field. The most important result of this communication is presented in Fig. 2a. The sample after cooling below T_C in zero external field aligns spontaneously with its full moment. Also, the direction of the sample magnetization is reversed as compared to the situation depicted in Fig. 2b (the peak appears in the $(--)$
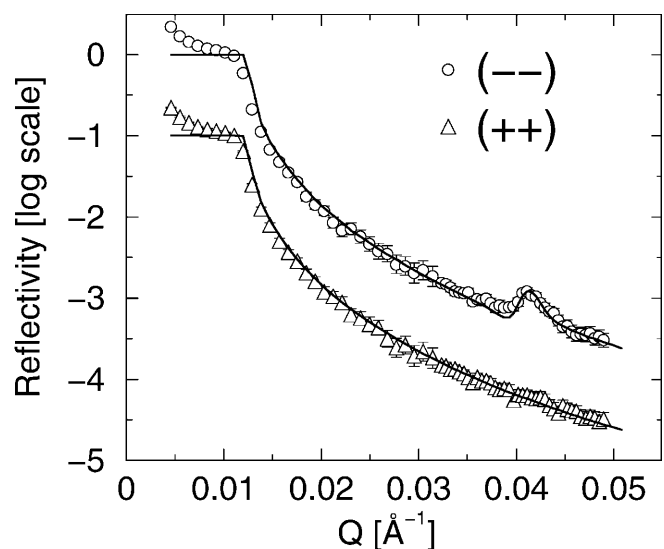

Table 1. Scattering length densities (SLDs) for GaMnAs and GaAs for the NSF and SF neutron scattering cross sections in 10^{-6} Å^{-2} units

GaMnAs $(++)$ $N(\bar{b} - \bar{p})$	GaMnAs $(--)$ $N(\bar{b} + \bar{p})$	GaMnAs $(+-)$ $N\bar{p}$	GaAs $N\bar{b}$
2.713	3.067	0.177	3.070

Fig. 1. Polarized NSF neutron reflectivity profiles for (50 ML/6 ML)×50 GaMnAs/GaAs superlattice taken at 2 G (stray) magnetic field. The data for the $(++)$ scattering process is shifted down by an order of magnitude for clarity. The presence of the SL peak in $(--)$ and not in $(++)$ indicates that the layer magnetization is opposite to the applied magnetic field
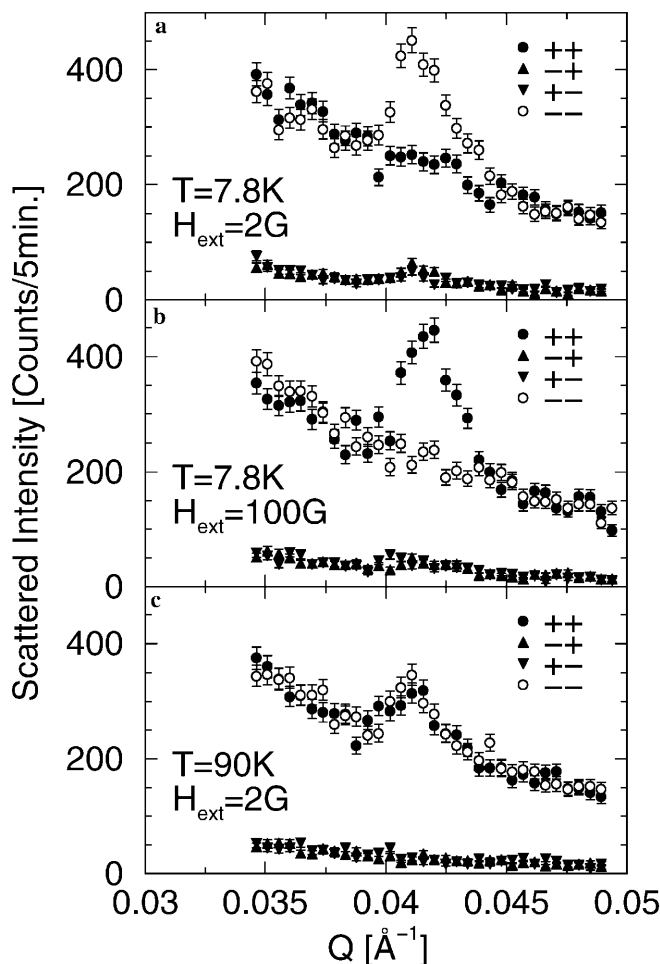
Fig. 2a–c. Polarized neutron diffraction profiles about the first-order SL Bragg peak for (50 ML/6 ML)×50 GaMnAs/GaAs superlattice. Non-spin-flip scattering processes (++) and (−−) as well as spin-flip ones (−+) and (+−) are presented. No peak in the spin-flip scattering indicates the absence of any horizontal component of the sample magnetization. Note the swap in the (++) and (−−) scattering (see (a) and (b)) after applying an external magnetic field of 100 G

NSF scattering process). Thus, the observed alignment cannot be caused by the residual stray fields (~ 2 G) originating from the 'guide-field', which has the same direction as the externally applied field. The height and the width of the SL peak are the same as for the sample saturated in an external field. Similarly, there is no spin-flip scattering present in this case. All these observations lead to the conclusion that each GaMnAs layer forms a single magnetic domain. Moreover, the magnetization of all layers is parallel, characteristic for ferromagnetically coupled superlattices.

In summary, our polarized neutron reflectivity experiments prove that the ferromagnetic order within a single GaMnAs layer is long-range. The magnetization of each individual layer is spontaneously saturated (single domain) and the spin of the Mn ion is very close to $\frac{5}{2}$, as can be inferred from the magnetic peak intensity (or the SLD contrast between GaMnAs and GaAs). That the magnetic SL Bragg peak appears at the same position as the structural one indicates that the ferromagnetic interlayer correlations exist in the investigated superlattices. Our last conclusion corroborates previous findings of interlayer coupling in GaMnAs/GaAs trilayer systems [11] based on magnetization hysteresis loop analysis.

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